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Carrier relaxation in InGaAs-GaAs quantum dots formed by activated alloy phase separation

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Abstract. Structures with InGaAs-GaAs quantum dots (QDs) formed by activated alloy phase separation have been studied by resonant photoluminescence. Near resonant PL at 20 K reveals a series of broad phonon replicas. This linewidth is explained by multiphonon relaxation processes involving LO phonons in the QDs, the wetting layer, interface, and GaAs as well as by additional scattering by acoustic phonons. The complex distribution of InGaAs in QDs formed by activated alloy phase separation is believed to cause a dispersion of QD phonon energies and to lead to a further broadening of phonon lines. For the sample with high QD confinement energy the photoluminescence lineshape depends on the excitation wavelength for near resonant excitation even at room temperature, which suggests a non-Fermi carrier distribution in the QD array.

Introduction

Carrier relaxation in quantum dots (QDs) has attracted much interest in recent years and been studied by different groups using resonant photoluminescence (PL) and PL excitation (PLE) spectroscopy [1–5]. Multi-phonon relaxation mechanisms [1, 2] as well as excited state absorption [3, 4] have been controversially proposed to explain PLE spectra of QD structures. In general, the relaxation processes are influenced by inhomogeneous broadening of QD array and competition between radiative and non-radiative recombination. Thus, prevalence of a concrete relaxation mechanism may depend on the parameters of a particular QD array. In Ref. [5] it has been shown that peaks due to excited state absorption as well as multiple LO resonance can be registered in the same PLE spectrum.

1. Experiment

InGaAs QDs were formed using the approach we refer to as Activated Alloy Phase Separation. In this growth approach a sheet of small Stranski–Krastanow islands was formed by depositing 2.1–2.7 ML of InAs and then overgrown by a 4–8 nm layer of $In_xGa_{1-x-y}Al_yAs$ ($x = 0.10–0.16$, $y = 0.0–0.15$). During the overgrowth, the strain field induced by each island causes migration of In atoms from the InGaAlAs alloy toward the islands leading to an increase in their size (Fig. 1). Details of the QDs growth as well as the structural and optical properties of these QDs have been described elsewhere [6–7]. A remarkable feature of QDs formed by the activated alloy phase separation is that their size and correspondingly confinement energies of the exciton in the ground state with respect to InGa(Al)As alloy and wetting layer can be tuned in a wide region 240–400 meV depending on the growth

Table 1.

Number	Initial islands	Overgrowth	QD size†	PL maximum (300 K)
QD1	2.1 ML InAs	4 nm $In_{0.1}Ga_{0.9}As$	12.5 nm	1.1 eV
QD2	2.5 ML InAs	5.5 nm $In_{0.13}Ga_{0.87}As$	15.5 nm	0.95 eV
QD3	2.5 ML InAs	8 nm $In_{0.15}Ga_{0.7}Al_{0.15}As$	17.5 nm	0.958 eV

† As determined by Transmission Electron Microscopy.

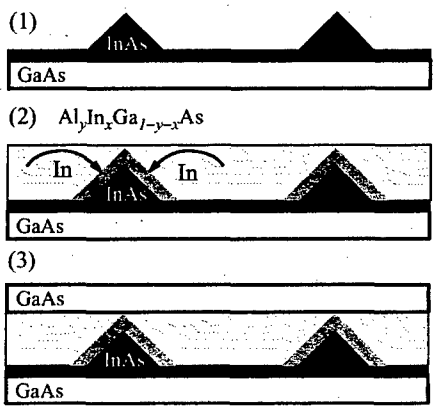

Fig. 1. A schematic diagram illustrating the Activated Alloy Phase Separation.

parameters [6–7]. In this paper we study three samples which we refer to as QD1–QD3. The growth parameters for the samples are presented in Table 1.

The photoluminescence (PL) was excited by an Ar^+ or Ti-sapphire laser (100 W/cm^2) and dispersed by a double-pass monochromator fitted with a Ge photodetector. The wavelength of the Ti-sapphire laser could be tuned in the region 900–1050 nm.

2. Results and discussion

To investigate the relaxation mechanisms in QDs we performed resonant PL studies using a Ti-sapphire laser. Figure 2 shows the resonant PL spectra for the structure QD2 at 10 K. When the energy of exciting photon is close to or above the InGaAs quantum well continuum energy, the shape of the PL spectrum depends only weakly on the excitation wavelength (Fig. 2(b)). By contrast, when the energy of exciting photon approaches the energy of the QD ground state transition (within 100–150 meV range) the shape of the PL spectrum changes and begins to depend significantly on the excitation wavelength. A set of equally-spaced peaks appears in the PL spectrum (Fig. 2(a)). The peaks are separated by a multiple of InAs- and GaAs-related LO-phonons with energies around 31 and 37 meV, respectively, and therefore we attribute the origin of these peaks to a multiple phonon relaxation mechanism. These resonant PL results are in an agreement with our previous PL excitation studies using lamp excitation [6–7].

In spite of the narrow spectral width of the excitation source and high resolution of the detection system, no sharp features were resolved for 2LO and 3LO replica for the sample QD2 pointing to an additional intrinsic broadening mechanism. Several reasons can account for the broadening of phonon lines. The first reason is a contribution of different phonons [2] including those from the InAs wetting layer (29.6 meV), InAs QD phonons (31.9 meV), interface phonons (35.0 meV) and GaAs phonons (36.6 meV). The scattering by acoustic phonons can also contribute to the broadening of phonon replicas. Taking into

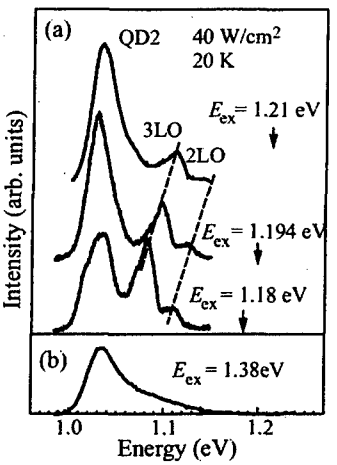

Fig. 2. PL spectra of the structure QD2 at 20 K under resonant (a) and non-resonant (b) excitation. The excitation wavelengths are indicated by arrows.

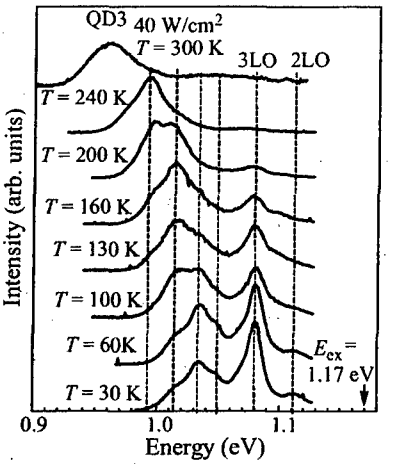

Fig. 3. Temperature dependence of the resonant PL spectra for the structure QD3.

account that the InGaAs LO phonon energy is altered by both In composition and strain, an additional reason for the broadening of phonon replicas may be caused by a complex distribution of strain and In concentration in the vicinity of the QDs formed by activated alloy phase separation.

Figure 3 shows the temperature dependence of the resonant PL spectra for the sample QD3. A set of peaks separated by a multiple of InAs- and GaAs-related LO-phonons from the laser line is observed at the resonant PL spectra of this structure, which is similar to the case of the structure QD2. For the non-resonant excitation PL peak shifts towards the low energies with the temperature increase, following the temperature dependence of the InAs bandgap energy. In contrast, the position of the peaks marked by the dashed lines in Fig. 3 is found to be temperature independent, which confirms that these lines are due to the multiple phonon relaxation mechanism.

For the structure QD1 the wavelength of the Ti-sapphire laser can be tuned closer to PL maximum, since the PL line of this sample is blue shifted as compared to those for the samples QD2 and QD3 (see Table 1). When the lasing wavelength is within the 50–30 meV range from the maximum of PL line recorded under non-resonant excitation several qualitatively new lines marked L and E appear in the spectra (Fig. 4). These lines are narrower than phonon replica in Fig. 2 and Fig. 3. The shift between line L and the excitation wavelength is still within the range of typical InGaAs LO phonon energies, whereas for line E this shift is about 43 meV. Thus, peak E might be due to the excited state absorption. Further experiments are in progress to verify the origin of lines L and E.

Depending on the sample temperature and the confinement of electron and hole energy levels with respect to the continuum, the carrier distribution within the QD array can be in equilibrium (Fermi) or non-equilibrium. At high temperatures or (and) low confinement energies the thermal nonequilibrium carrier emission, lateral transport via wetting layer and matrix and then recapture results in the quasi-equilibrium distribution of carriers in the array. At low temperatures or (and) efficient confinement of electron and hole energy levels, the carrier escape from the individual QDs is suppressed and the QD array can not be described by a common quasi-Fermi level.

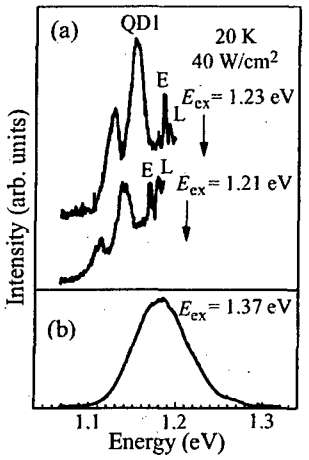

Fig. 4. PL spectra of the structure QD1 at 20 K under resonant (a) and non-resonant (b) excitation. The excitation wavelengths are indicated by arrows.

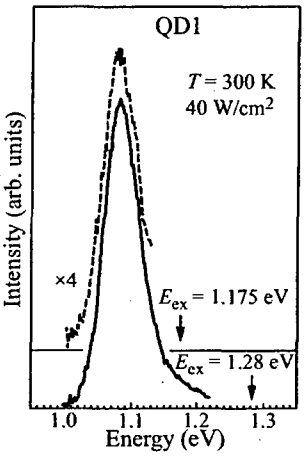

Fig. 5. PL spectra of the structure QD1 at 300 K for different excitation wavelength.

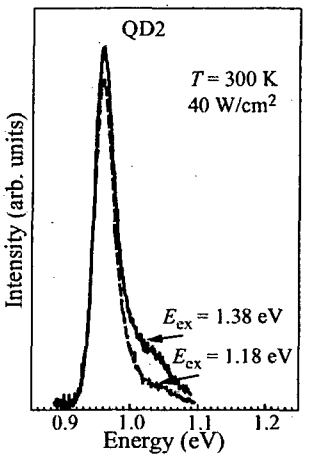

Fig. 6. PL spectra of the structure QD2 at 300 K for different excitation wavelength.

At 20 K lineshape for the samples QD1–QD3 dramatically depends on the excitation wavelength under resonant excitation (Fig. 2–4), which suggests that at 20 K there is no lateral transport and correspondingly quasi-Fermi level in these QDs arrays. However at room temperature the shapes of resonant PL spectra for the sample QD1 does no longer seem to depend on the excitation wavelength (Fig. 5). Thus, for this sample a common quasi-Fermi level is already established at 300 K. In contrast for the structure QD2 PL line shape depends on the excitation wavelength even at room temperature (Fig. 6). We note that for this sample the exciton confinement energy is higher than that for the sample QD1 (see Table 1). The data shown in Fig. 6 suggest a non-thermal distribution of electrons and holes at room temperature for the sample QD2 at room temperature.

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